Supporting Information

Probing the Intrinsic Properties of Exfoliated Graphene: Raman Spectroscopy of Free-Standing Monolayers

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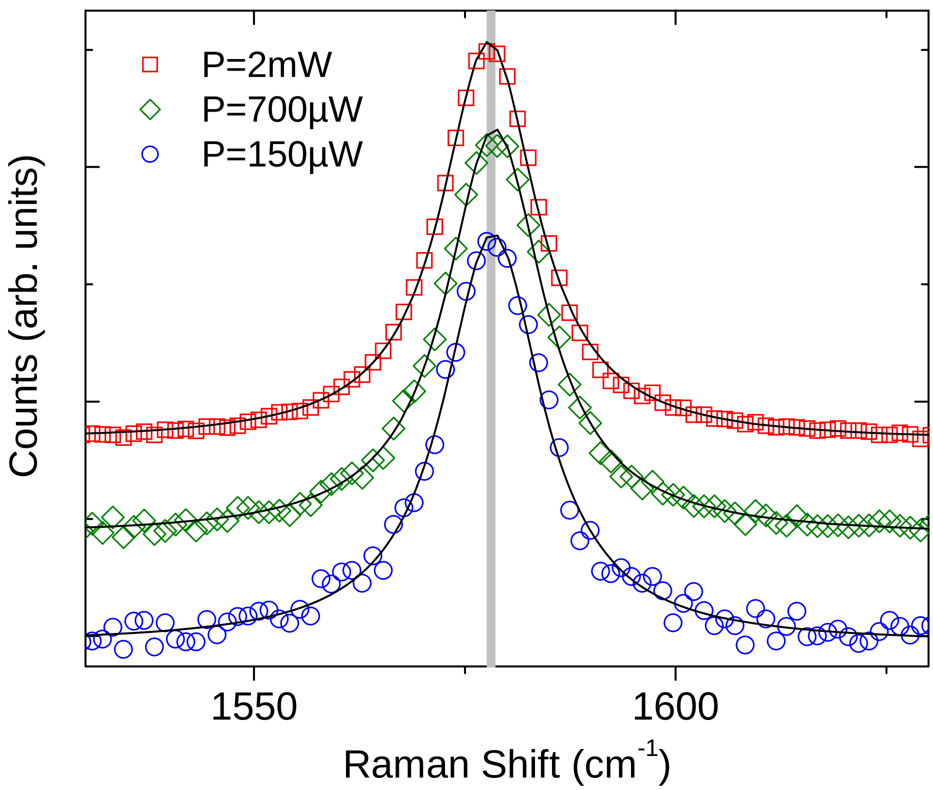

Figure S1: High-resolution G-mode Raman spectra measured on a single diffraction limited spot of a free-standing graphene monolayer for laser powers of 150µW (dots), 700µW (diamonds), and 2mW (squares). The solid lines are Lorentzian fits. Within experimental resolution, no shifts or changes in line shape are observed. The G-mode frequency and width are 1578±0.5cm^{-1} and 14±0.5 cm^{-1}, respectively.

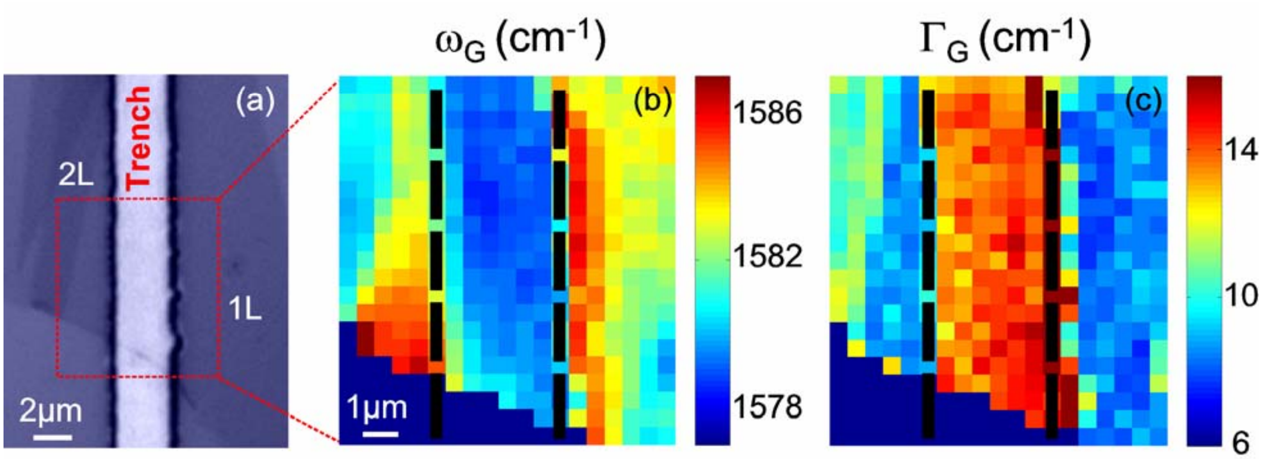

Figure S2: Raman maps of a large-area ($9\times10\ \mu m^2$) graphene monolayer, showing the spatial homogeneity of the free-standing portion. Maps of the G-mode frequency (b) and width (c) were recorded over the boxed region of the sample shown in the optical micrograph (a). The two dashed black lines in (b) and (c) designate the borders of the trench. Data in (b) and (c) are deduced from Voigt fits. On the free-standing region, we find mean values of $\langle\omega_G\rangle$=1579.5 cm^{-1} and $\langle\Gamma_G\rangle$=14 cm^{-1}, indicative of the intrinsic character of graphene sample (see manuscript). In addition, the corresponding standard deviations of 0.5cm^{-1} and 1cm^{-1} highlight the high spatial homogeneity of the suspended portion of the sample.

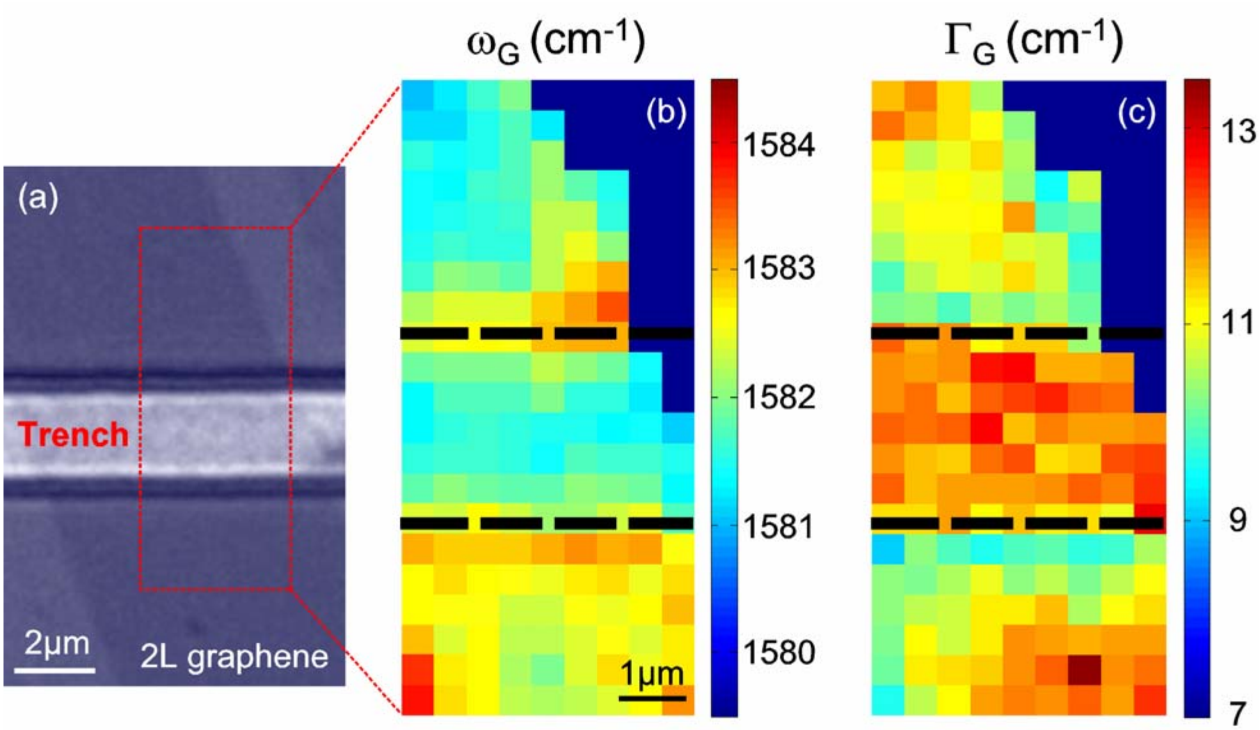

Figure S3: Raman mapping of a graphene bilayer sample. The scanned area, which includes both free-standing and supported regions, is shown in the optical micrograph of (a). The G-mode frequency and width are displayed in (b) and (c). The two dashed black lines in (b) and (c) designate the borders of the trench. Data in (b) and (c) are deduced from Voigt fits. On the suspended region, we find mean values of $\langle\omega_G\rangle$=1581.5±0.5 cm^{-1} and $\langle\Gamma_G\rangle$=12±1 cm^{-1}.

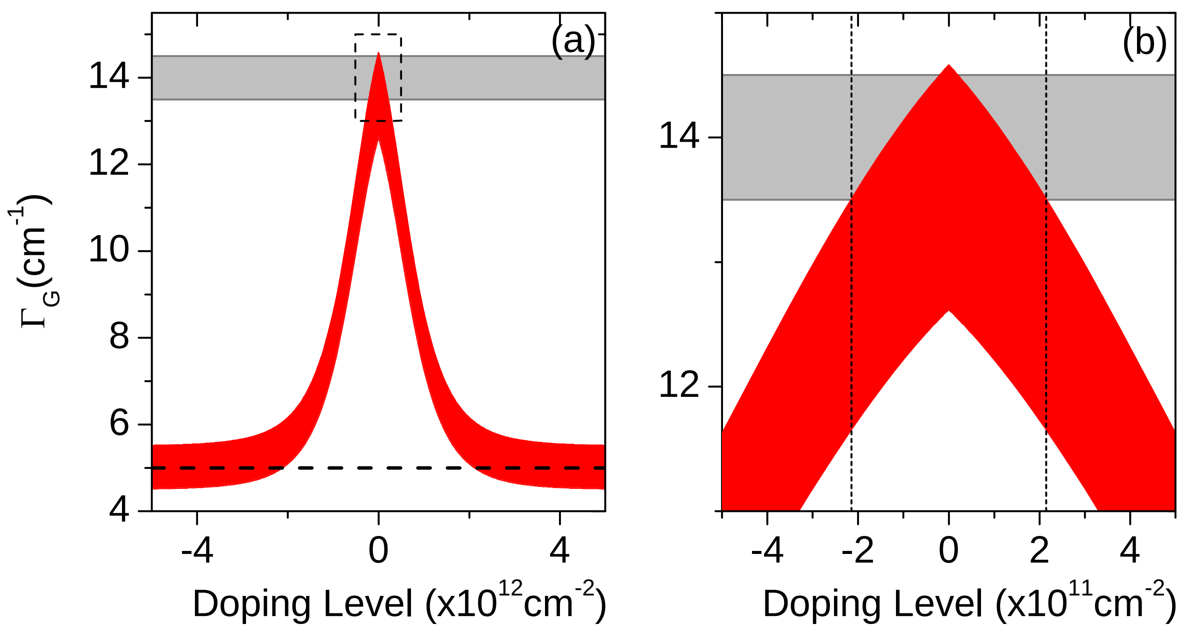

Figure S4: (a) Comparison between the experimental value of width of the Raman G-mode, Γ_G, on a free-standing graphene monolayer (14±0.5cm^{-1}, grey area) and simulations based on Eq. 1 (red area). We use our experimentally determined upper bound for width, Γ_0, not arising from electron-hole pair generation (5±0.5 cm^{-1}, dashed black line) and the mean value of electronic contribution, $\Delta\Gamma$, (9±0.5 cm^{-1}) measured on graphene FETs[1-4]. The red area accounts for the variation of the experimental parameters used in the simulations. (b) Expanded view of the boxed region in (a). An upper bound of ~2×10^{11} cm^{-2} (see dashed vertical lines) for the doping level is deduced.

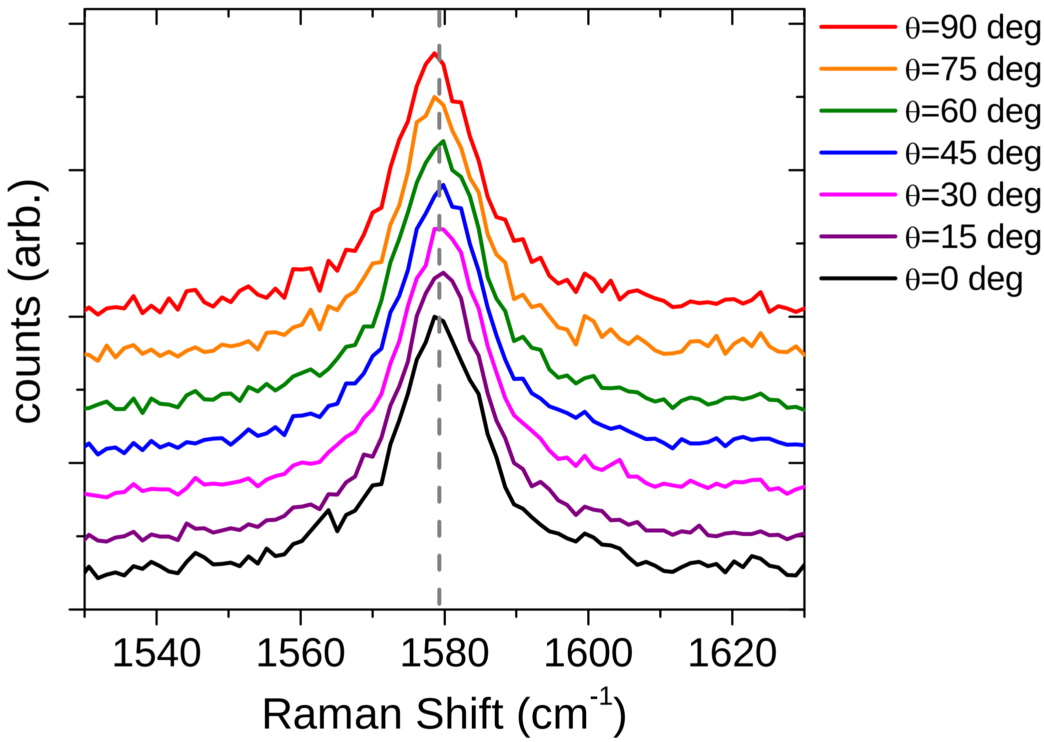

Figure S5: High-resolution spectra of the Raman G mode measured from a single diffraction limited spot of a free-standing graphene monolayer as a function of the polarization of the detected scattered light. The excitation beam was of fixed linear polarization. The angle θ is given with respect to the orientation of the pump polarization. The data were recorded using a laser power of 700μW. The spectra are normalized and have been offset for clarity. We see neither frequency shifts nor changes in the line shape of the feature. Using Lorentzian fits, we obtain a frequency of 1579.5 ± 0.5cm^{-1}and a width of 13.5 ± 0.5 cm^{-1} for this feature.

References

1. Yan, J.; Zhang, Y.; Kim, P.; Pinczuk, A. *Physical Review Letters* 2007, 98, 166802.
2. Pisana, S.; Lazzeri, M.; Casiraghi, C.; Novoselov, K. S.; Geim, A. K.; Ferrari, A. C.; Mauri, F. *Nature Materials* 2007, 6, 198.
3. Das, A.; Pisana, S.; Chakraborty, B.; Piscanec, S.; Saha, S. K.; Waghmare, U. V.; Novoselov, K. S.; Krishnamurthy, H. R.; Geim, A. K.; Sood, A. K.; Ferrari, A. C. *Nature Nanotechnology* 2008, 3, 210.
4. Das, A.; Chakraborty, B.; Piscanec, S.; Pisana, S.; Sood, A. K.; Ferrari, A. C. *ArXiv:0807.1631* 2008.